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High Energy Density Interpenetrating Networks from Ionic Networks and Silicone

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ABSTRACT

The energy density of dielectric elastomers (DEs) is sought increased for better exploitation of the DE technology since an increased energy density means that the driving voltage for a certain strain can be lowered in actuation mode or alternatively that more energy can be harvested in generator mode. One way to increase the energy density is to increase dielectric permittivity of the elastomer. A novel silicone elastomer system with high dielectric permittivity was prepared through the development of interpenetrating networks from ionically assembled silicone polymers and covalently crosslinked silicones. The system has many degrees of freedom since the ionic network is formed from two polymers (amine and carboxylic acid functional, respectively) of which the chain lengths can be varied, as well as the covalent silicone elastomer with many degrees of freedom arising from amongst many the varying content of silica particles. A parameter study is performed to elucidate which compositions are most favorable for the use as dielectric elastomers. The elastomers were furthermore shown to be self-repairing upon electrical breakdown.

Keywords: Silicone elastomers, dielectric elastomers, PDMS, self-repairing, ionic moieties

1. INTRODUCTION

Dielectric elastomers (DEs) are based on an elastomer film sandwiched between thin and compliant electrodes. This emerging technology can be used for actuators, sensors and generators due to their ability to expand and contract under an applied electric field or to generate electrical energy when subjected to mechanical motion. High-dielectric permittivity elastomers are required for improving the performance of many of the current devices based on DEs so that either a reduced driving voltage or an increased actuation at a given voltage can be obtained. The high driving voltages which are necessary to activate the DE (usually in the order of several kVs) put some limitations on the applicability of the technology due to overly expensive electronics. The driving voltages of DEs are typically lowered by one of two methods, namely either – with the strongest effect – by reducing the elastomer thickness (d) and/or – with less impact – by increasing the ratio between the dielectric permittivity (ε') and the Young's modulus (Y). For many elastomers it is difficult to go below the commonly applied thickness of 20 μm due to processing issues such as the release of thin films from the curing substrate.[1]

Several approaches to increasing dielectric permittivity of DEs have been investigated, including encapsulation of conductive fillers[2], grafting of dipoles[3–6] and addition of high-permittivity fillers.[7,8] Many of these systems do, however, fail in increasing the actuation performance (ε'/Y) or the maximum achievable actuation ($\varepsilon'/Y*E_B^2$, where E_B is the dielectric breakdown strength) since the addition of hard additives, whether grafted or not, often increases Young's modulus to the same extent as the dielectric permittivity is increased and furthermore the breakdown strength may also be significantly lowered. Moreover, in order for an elastomer to be applicable as a DE many other requirements have to be fulfilled such as fast response time, high mechanical stability and very low conductivity.[9]

The focus of this work is to develop very high dielectric permittivity elastomers such that a decrease in electrical breakdown strength can be tolerated and an improvement in the actuation performance is still obtained.

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Recently a novel class of polymers was introduced, namely the so-called ionic networks[10–12] which are based on macromolecular assembly through ionic interactions. These materials have been shown to possess extremely high dielectric permittivities and in this work we explore the resulting properties of interpenetrating networks of silicone with siloxane based ionic networks. Pei et al[13–15] have previously explored interpenetrating networks as a means of maintaining prestretch in acrylics based elastomers. Here we utilize interpenetrating networks to maintain a covalently bonded network throughout the sample as well as to dilute the slightly conductive nature of the ionic networks since pure ionic networks have conductivities of around 10^{-9} S/cm compared to 10^{-13} S/cm for common RTV/LSR elastomers.

2. EXPERIMENTAL

2.1 Materials and methods

6-7% Aminopropylmethylsiloxane dimethylsiloxane copolymer AMS-162 ($\bar{M}_w \approx 4500$ g mol^{-1}), carboxydecyl terminated poly(dimethyl siloxane) DMS-B12 ($\bar{M}_w = 1000$ g mol^{-1}), DMS-B25 ($\bar{M}_w = 10{,}000$ g mol^{-1}), vinyl-terminated PDMS (DMS-V35) ($\bar{M}_w = 49{,}500$ g mol^{-1}) and tetrakis(dimethylsiloxy)silane (tetra-functional cross-linker) ($\bar{M}_w = 329$ g mol^{-1}) were obtained from Gelest Inc. The commercial silicone elastomers ELASTOSIL® RT625, POWERSIL® XLR630 and ELASTOSIL® LR3043/30 were acquired from Wacker Chemie AG, and all are two-component silicone rubbers; RT625 is a room-temperature vulcanisation rubber (RTV), where the mixing ratio of parts A and B are 9:1. XLR630 and LR3043/30 are liquid silicone rubbers (LSRs), where the mixing ratios of parts A and B are 1:1. The platinum cyclovinylmethyl-siloxane complex catalyst (511) was supplied by Hanse Chemie AG. The silica particles (hexamethyldisilazane treated amorphous silicon dioxide) were purchased from Fluorochem. The compositions of samples are shown in Table 1.

Table 1. Component concentrations of pure PDMS and commercial silicone elastomers, as well as prepared ionic and interpenetrating networks.

#		Networks	Silicone [wt%]	Ionic network [wt%]	SiO_2 [wt%]
1	**Pure PDMS**	PDMS V35	100	0	0
2	**Commercial silicones**	RT625	73	0	27
3		XLR630	63	0	37
4		LR3043/30	27	0	73
5	**Ionic networks**	AMS162+B12	0	100	0
6		(AMS162+B12) : SiO_2 = 80wt.% : 20wt.%	0	80	20
7		(AMS162+B12) : SiO_2 = 70wt.% : 30wt.%	0	70	30
8	**Interpenetrating networks**	XLR630 : (AMS162+B12) = 70wt.% : 30wt.%	44.1	30	25.9
9		RT625 : (AMS162+B25) = 50wt.% : 50wt.%	36.5	50	13.5
10		RT625 : (AMS162+B25) : SiO_2 = 50wt.% : 50wt.% : 10wt.%	32.8	45	22.2
11		LR3043/30 : (AMS162+B12) = 70wt.% : 30wt.%	18.9	30	51.1
12		LR3043/30 : (AMS162+B12) = 50wt.% : 50wt.%	13.5	50	36.5
13		LR3043/30 : (AMS162+B12) = 30wt.% : 70wt.%	8.1	70	21.9
14		LR3043/30 : (AMS162+B12) : SiO_2 = 30wt.% : 70wt.% : 10wt.%	7.3	63	29.7
15		LR3043/30 : (AMS162+B12) = 10wt.% : 90wt.%	2.7	90	7.3

Dielectric relaxation spectroscopy (DRS) was performed on a Novocontrol Alpha-A high-performance frequency analyser (Novocontrol Technologies GmbH & Co. KG, Germany) operating in the frequency range 10^{-1}-10^{6} Hz at 23°C. The sample diameters were 25 mm, while thicknesses were approximately 0.5-1.0 mm. Electrical breakdown tests were performed on an in-house-built device based on international standards (IEC 60243-1 (1998) and IEC 60243-2 (2001)). Film thicknesses were measured through the microscopy of cross-sectional cuts, and the distance between the spherical electrodes was set accordingly with a micrometer stage

and gauge. An indent of less than 5% of sample thickness was added, to ensure that the spheres were in contact with the sample. The polymer film was slid between the two spherical electrodes (radius of 20 mm), and the breakdown was measured at the point of contact with a stepwise increasing voltage applied (50-100 V/step) at a rate of 0.5-1 steps/s. Each sample was subjected to 12 breakdown measurements, and an average of these values is given as the breakdown strength of the sample. The sample thicknesses were approximately 40-100 μm.

The initial silica concentration in the commercial silicone elastomers was evaluated by thermal gravimetric analysis (TGA) on a Q500 from TA Instruments, and measurements were carried out under a nitrogen atmosphere at a heating rate of $10^{o}C\ min^{-1}$ from room temperature to $900^{o}C$. In commercial silicone formulations the majority of residue at 900°C corresponds to the initial silica concentration[16].

Self-healing properties were evaluated by marking samples with a dark marker and cutting the samples in two pieces. The samples were then assembled in two configurations, namely head to head or back to back (with head being the freshly cut part).

2.2 General procedure for elastomer synthesis

Generic sample preparation is described below. All ionic networks were prepared by mixing stoichiometric amounts of amine-functional PDMS (AMS-162) and carboxylic acid-functional PDMS (DMS-B12 or DMS-B25) using Speedmixer (DAC 150FVZ, Hauschild Co.) for 2 minutes at 2000 rpm. Subsequently commercial two-component silicone mixtures were mixed into the above ionic networks using Speedmixer for 2 minutes at 2000 rpm. Finally silica fillers were mixed using the Speedmixer for 2 minutes at 2000 rpm for the filled samples.

Pure PDMS reference is reacted from a stoichiometric ratio of components. The concentration of platinum catalyst is 1.6 ppm. The mixture was mixed using Speedmixer for 2 minutes at 2000 rpm.

The uniform mixtures were cast with a film applicator (3540 bird, Elcometer, Germany) on a glass substrate and fully cured after 24h at 100 ^{o}C in the oven.

3. RESULTS AND DISCUSSION

Dielectric interpenetrating networks were prepared according to Figure 1. The networks were prepared by mixing carboxydecyl-terminated PDMS of either 1000 g mol^{-1} (DMS-B12) or 10,000 g mol^{-1} (DMS-B25) with amino-functional PDMS (AMS-162). These components upon protonation of the amino-groups formed ionic bonds and thereby formed the ionic part of the interpenetrating silicone network (Figure 2). The ionic networks were hereafter mixed with various commercial silicone elastomer systems in order to elucidate the effect of the different formulations on the final dielectric and breakdown properties of the interpenetrating networks. The commercial two-component systems were respectively: RT625, an RTV silicone, and XLR630 and LR3043/30 both LSR silicones. Furthermore, some samples were mixed with additional silica particle to reinforce systems with high concentrations of the ionic networks.

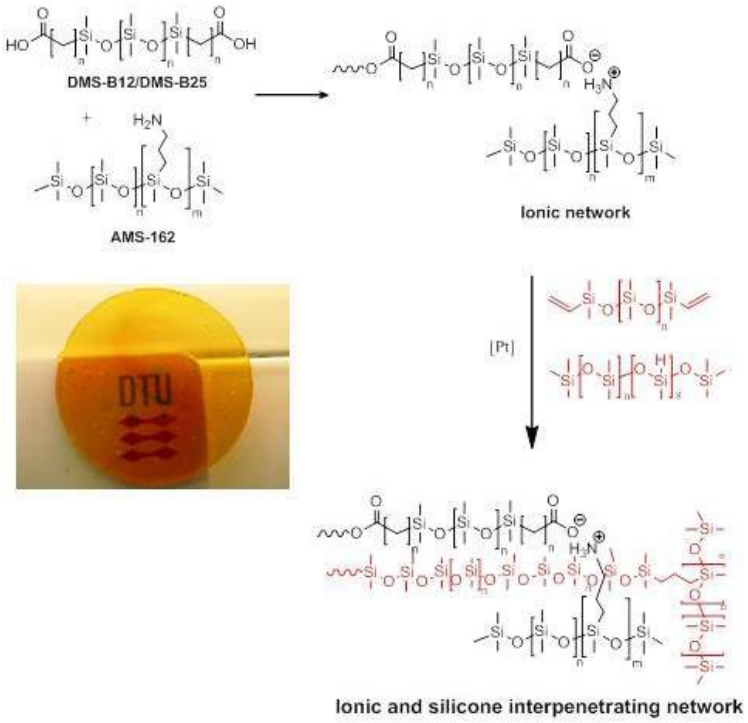

Figure 1. Reaction for preparation of interpenetrating network from ionic and hydrosilylation networks.

Figure 2. The reactants (a) before and (b) after heating at 115°C for 6 days. (c) A pure ionic network.

The effect of the concentration and type of commercial silicone system on the dielectric properties of the interpenetrating networks was determined by dielectric relaxation spectroscopy on discs of approximately 0.5 to 1 mm thick films. The resulting dielectric permittivities as function of composition at two different frequencies (0.1 Hz and 1 MHz) are shown in Figure 3 and are furthermore summarized in Table 2.

Table 2. Summary of dielectric permittivity (ε_r) and loss tangent (tan δ) at 0.1 Hz and 1 MHz, breakdown strengths (E_B), Young's moduli and loss (tan δ), as well as the figure of merit calculations.

#		Networks	Breakdown strength	Dielectric properties				Viscoelastic properties		Figure of merit			
			E_B	ε_r	ε_r	tan δ	tan δ	Y=3G'	tan δ	F_{om} (DEA)	F_{om} (DEA)	F_{om} (DEG)	F_{om} (DEG)
				0.1Hz	1MHz	0.1Hz	1MHz	0.01Hz	0.01Hz	0.1Hz	1MHz	0.1Hz	1MHz
			[V/μm]					[kPa]					
1	**Pure PDMS**	PDMS V35	40	2.5	2.5	0.130	4.1×10^{-4}	64.3	0.06	0.25	0.25	0.19	0.19
2	**Commercial silicones**	RT625	87	2.8	2.8	0.189	4.2×10^{-4}	84.4	0.03	1	1	1	1
3		XLR630	120	3.2	3.2	0.299	5.5×10^{-4}	227.2	0.04	0.79	0.79	2.13	2.13
4		LR3043/30	130	2.9	2.9	0.190	5.5×10^{-4}	253.3	0.08	0.77	0.77	2.30	2.30
5	**Ionic networks**	AMS162+B12	72	7.5×10^{3}	6.2	4.76	0.199	37.3	0.01	4107	3.41	1813	1.50
6		(AMS162+B12) : SiO_2 = 80wt.% : 20wt.%	83	4.5×10^{3}	3.7	3.34	0.147	47.0	0.07	2620	2.15	1460	1.20
7		(AMS162+B12) : SiO_2 = 70wt.% : 30wt.%	101	3.1×10^{3}	3.4	2.87	0.133	124.2	1.74	1013	1.10	1491	1.62
8	**Interpenetrating networks**	XLR630 : (AMS162+B12) = 70wt.% : 30wt.%	69	6.7	2.8	5.51	0.042	37.0	0.36	3.39	1.41	1.48	0.62
9		RT625 : (AMS162+B25) = 50wt.% : 50wt.%	76	9.8	4.1	3.97	0.029	61.4	0.09	3.62	1.51	2.63	1.10
10		RT625 : (AMS162+B25) : SiO_2 = 50wt.% : 50wt.% : 10wt.%	82	13.3	5.0	4.21	0.029	70.5	0.12	5.00	1.89	4.17	1.58
11		LR3043/30 : (AMS162+B12) = 70wt.% : 30wt.%	45	13.0	3.4	3.38	0.032	255.1	0.10	0.41	0.11	1.23	0.32
12		LR3043/30 : (AMS162+B12) = 50wt.% : 50wt.%	78	1.7×10^{2}	3.8	11.6	0.140	113.7	0.08	24.0	0.80	33	1.07
13		LR3043/30 : (AMS162+B12) = 30wt.% : 70wt.%	90	1.5×10^{3}	3.8	4.44	0.136	30.9	0.05	1579	3.93	579	1.44
14		LR3043/30 : (AMS162+B12) : SiO_2 = 30wt.% : 70wt.% : 10wt.%	98	6.4×10^{2}	3.4	2.59	0.093	157.3	0.05	153	0.83	285	1.54
15		LR3043/30 : (AMS162+B12) = 10wt.% : 90wt.%	87	2.1×10^{3}	4.1	2.3	0.076	30.5	0.03	2008	4.05	724	1.46

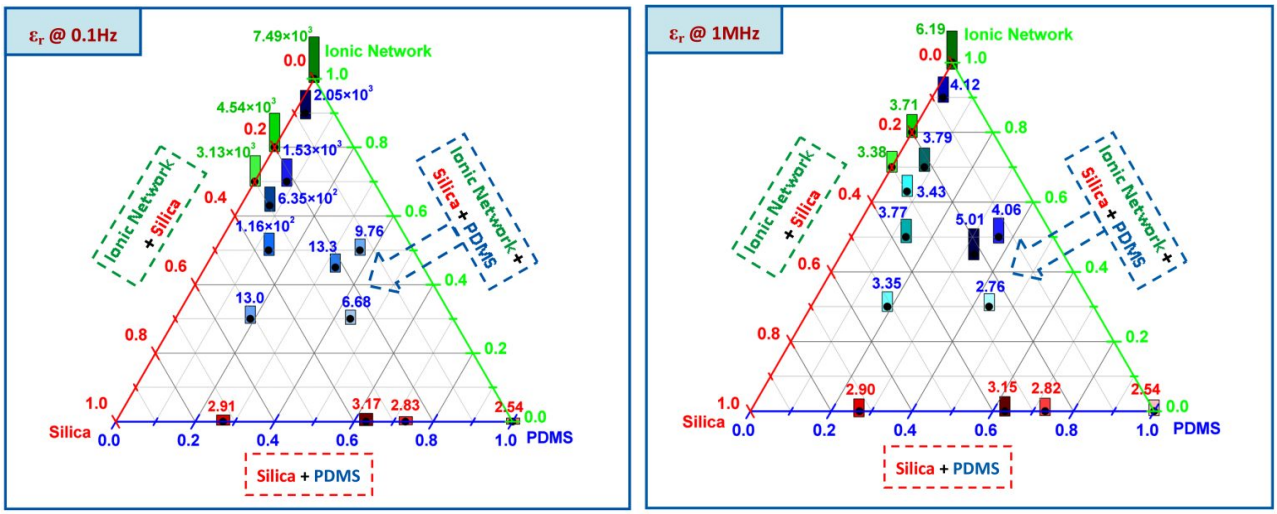

Figure 3. Dielectric permittivity as function of composition of interpenetrating ionic and silicone networks at 0.1 Hz and 1 MHz, respectively.

As seen in the ternary plots in Figure 3 pure PDMS networks have dielectric permittivities of approximately ε_r = 2.5-3.2 (red numbers) independent of frequency. Pure ionic networks made from carboxydecyl-terminated PDMS and amino-functional PDMS and different concentrations of reinforcing silica particles display very high dielectric permittivities at low frequency (as high as $\varepsilon_r = 7.5\times10^3$ for pure ionic network (green numbers)). At higher frequency, the pure ionic network also has higher dielectric permittivity than pure silicone (ε_r = 6.2), which corresponds to a two-fold increase compared to pure silicone. The blue numbers in the ternary plots corresponds to the measured dielectric permittivities of the interpenetrating ionic and silicone networks. As seen in Figure 3, the dielectric permittivities of the interpenetrating networks are very high at low frequencies ranging from $\varepsilon_r = 6.7\text{-}2\times10^3$). At higher frequencies the highest dielectric permittivity of ε_r = 5 was obtained for a network with 50% ionic network and 50% silicone network (sample 10). Higher concentrations of ionic network did not necessarily induce a higher dielectric permittivity at higher frequencies. The dielectric permittivity only increases at low frequencies with increasing concentration of ionic network.

The Young's moduli of the samples were determined by linear rheology where the Young's modulus is calculated from the measured storage modulus, G', since Y = 2(1 + ν)G, where G = G'(ω → 0). Due to the incompressibility of silicones (Poisson's ratio (ν) close to 0.5) the Young's modulus can be obtained as Y = 3G', which describes frequency-independent elastic moduli up to a strain of approximately 15%. Furthermore, highly frequency-dependent viscous loss can be determined from the measured loss modulus, G'', and is usually referred to via the loss tangent, tan δ = G''/G'. The Young's modulus calculated from G' at 0.01 Hz and the loss as tan δ are shown in Table 2. For pure PDMS and the commercial silicones it can be seen that Young's modulus increases with increasing concentration of reinforcing silica particles. The particles, therefore, greatly increases the stiffness of the silicone networks. Furthermore, the viscous losses, tan δ, are low for all these samples meaning that they are well cross-linked. The pure ionic network (sample 5) has significantly lower Young's modulus and is therefore a lot softer than the silicone elastomers. The viscous loss is, however, as low as for the silicone elastomers which mean that the ionic network is very well cross-linked. Upon addition of silica particles (sample 6 and 7) the Young's modulus of the ionic networks increases and at 30 wt% silica particles the ionic network has become significantly stiffer. The viscous loss for this sample is however high (>1) which indicates that the addition of high amounts of silica particles to the ionic networks to some extent hinders the cross-linking reaction. The Young's moduli of the interpenetrating networks vary depending of the type of commercial silicone elastomer used and the addition of silica particles. For the XLR630 elastomer the addition of 30wt% ionic network decreases the Young's modulus significantly from 227.2 kPa to 37.0 kPa. The viscous loss is however increased considerably although it remains <1. The Young's modulus of the RT625 silicone elastomers is only decreased from 84.4 kPa to 61.4 kPa upon addition of 50wt% ionic network (sample 9) and the viscous loss remains low meaning that

the mechanical integrity of this sample is maintained. Adding silica particles to this elastomer (sample 10) increases the modulus somewhat to 70.5 kPa and increases the viscous loss ten-fold indicating increased damping behaviour. Again this could be due to the silica particles hindering the cross-linking reaction of the ionic networks. For the LR3043/30 silicone elastomer the addition of 30wt% ionic network does not alter the Young's modulus. The viscous loss is however increased. Increasing the concentration of ionic network in this elastomer to 50wt%, 70wt% and 90wt% decreases the Young's modulus significantly (from 253.3 kPa for pure LR3043/30 to 113.7 kPa, 30.9 kPa and 30.5 kPa for 50, 70 and 90 wt% ionic network, respectively). Common for all these samples is that the viscous losses remain low even at very high concentration of ionic network. The addition of 10wt% silica particles to an elastomer with 70wt% ionic network (sample 14) increases the Young's modulus significantly (from 30.9 kPa to 157 kPa) while still maintaining low viscous loss. The viscoelastic measurements indicate that the RT625 and LR3043/30 elastomers are more suitable for the preparation of ionic interpenetrating networks compared to XLR630 since they maintain low viscous losses upon addition of ionic network. Furthermore, LR3043/30 can be used with reinforcing silica particles without the losses being compromised.

The measured dielectric breakdown strength for the interpenetrating and pure networks is shown in Figure 4. As seen in red numbers in the bottom right of Figure 4, the breakdown strength of pure unreinforced PDMS is as low as 40 V/µm. The dielectric breakdown strength then increases with increasing concentration of silica particles reaching 130 V/µm for LR3043/30. This is due to increasing stiffness which has a large influence on the breakdown of DEs since increased stiffness supresses electromechanical instability[17]. The breakdown strength of pure ionic networks also increases with increasing concentration of silica particles from 72 V/µm for a pure ionic network to 101 V/µm for a particle filled network with 30 wt% silica. The breakdown strengths of the interpenetrating networks range from 45 V/µm to 90 V/µm and therefore in all cases remain higher than that of a pure PDMS network.

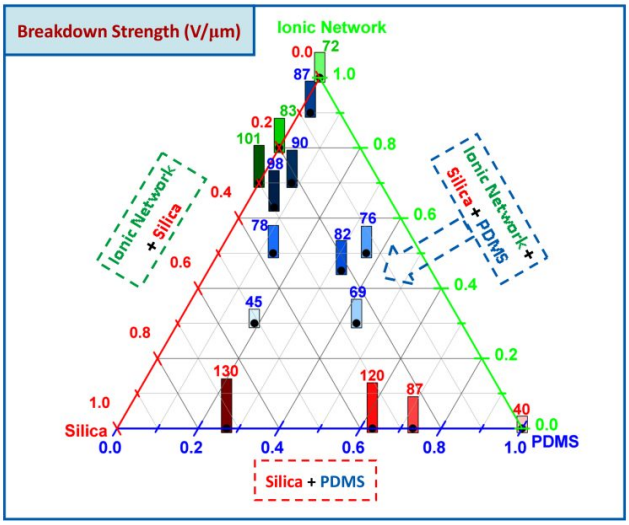

Figure 4. Dielectric breakdown strengths as function of network composition.

The equations for figure of merits (F_{om}) to assess elastomer performance as a dielectric elastomer actuator (DEA) and a dielectric elastomer generator (DEG) are defined as follows[7]:

$$F_{om}(\text{DEA}) = \frac{3\varepsilon_r\varepsilon_0 E_{breakdown}^2}{Y} \quad (1)$$

$$F_{om}(\text{DEG}) = \frac{\varepsilon_r\varepsilon_0 E_{breakdown}^2}{2\varphi} \quad (2)$$

where E_B (V/μm) is the electrical field at which electrical breakdown occurs, ε_r the relative dielectric permittivity, ε_o the permittivity of free space (8.85×10^{-12} F/m) and φ the strain energy function of the elastomer, which is assumed to be equivalent for each silicone formulation. The figure of merit is calculated relative to RT625 which is used as a reference material and therefore has $F_{om} = 1$. The results of the calculations are shown in Figure 5 and summarised in Table 2.

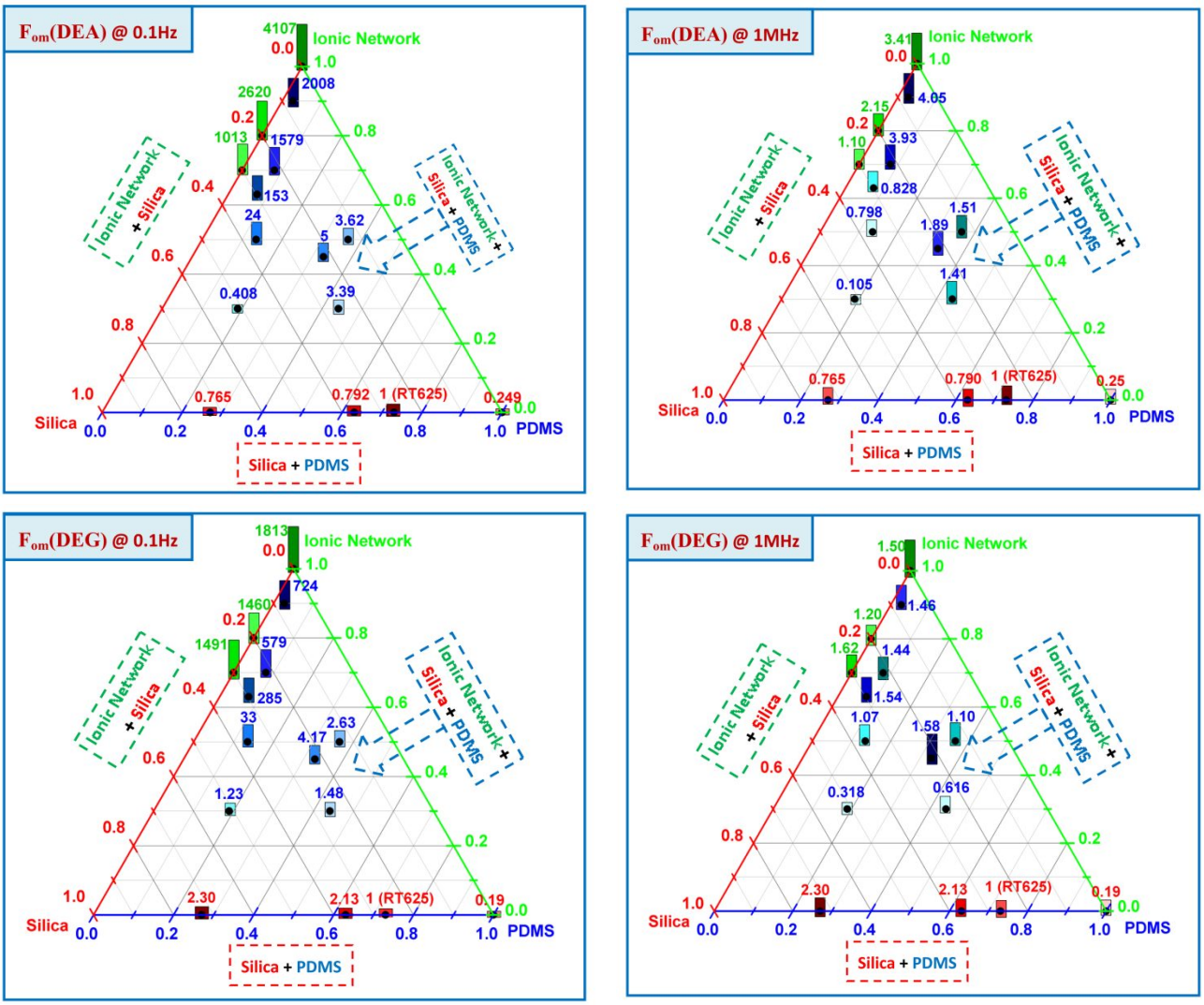

Figure 5. Relative figure of merit calculations for DEAs and DEGs at 0.1 Hz and 1 MHz.

The relative figure of merit calculations for a DEA illustrate that at both 0.1 Hz and 1 MHz sample 13 and 15, which are prepared with the commercial elastomer LR3043/30, show the highest overall improvement in predicted actuator properties. This is due to the relatively soft nature of these films and increased dielectric permittivity. Furthermore a pure ionic network without reinforcing silica (sample 5) also has very high relative figure of merit at low frequencies due to its very high dielectric permittivity at this frequency. This sample, however, has reduced dielectric breakdown strength. The predicted properties for a DEG at 0.1 Hz have improved the most for the samples prepared with pure ionic network (samples 5-7). The interpenetrating networks prepared with LR3043/30 and high concentration of ionic networks (sample 13-15) also show high increases in overall properties due to the very high obtained dielectric permittivities at this frequency. At higher frequency (1 MHz) the relative figures of merit and thereby the overall DEG properties have

not increased much compared to the RT625 reference sample. Therefore the most promising use of the ionic and interpenetrating networks based on the figure of merit calculations would be as DEAs or DEGs run at low frequencies.

The samples were shown to be self-healing upon cutting, i.e. a mechanical disruption. As shown by Yu and Skov traditional silicones are not capable of growing together when the elastomer is prepared in stoichiometric amounts[16]. However, the ionic networks are capable of reforming again and the resulting network is capable of supporting more than 100% elongation of the reassembled samples. This is illustrated in Figure 6.

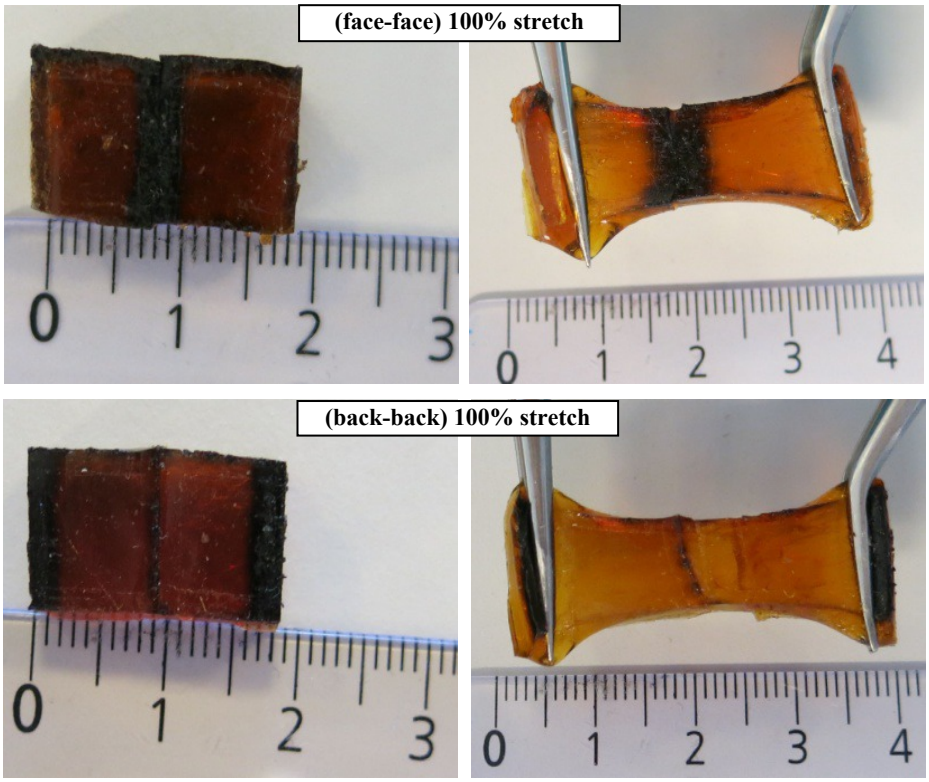

Figure 6. Self-healing properties of the silicone elastomers. Both face-face and back-back configurations could be extended to more than 100% before breaking. The dark areas are from the cutting line.

Subsequently the samples were subjected to electrical breakdowns by applying a destructive electrical field to the samples. The samples could also repair themselves after this mode of failure (Figure 7). This is a very promising feature of this type of networks since it would potentially be a way to increase life-time of DEs if the electro-mechanical failure does not lead to tearing of films but solely leads to puncture voids etc.

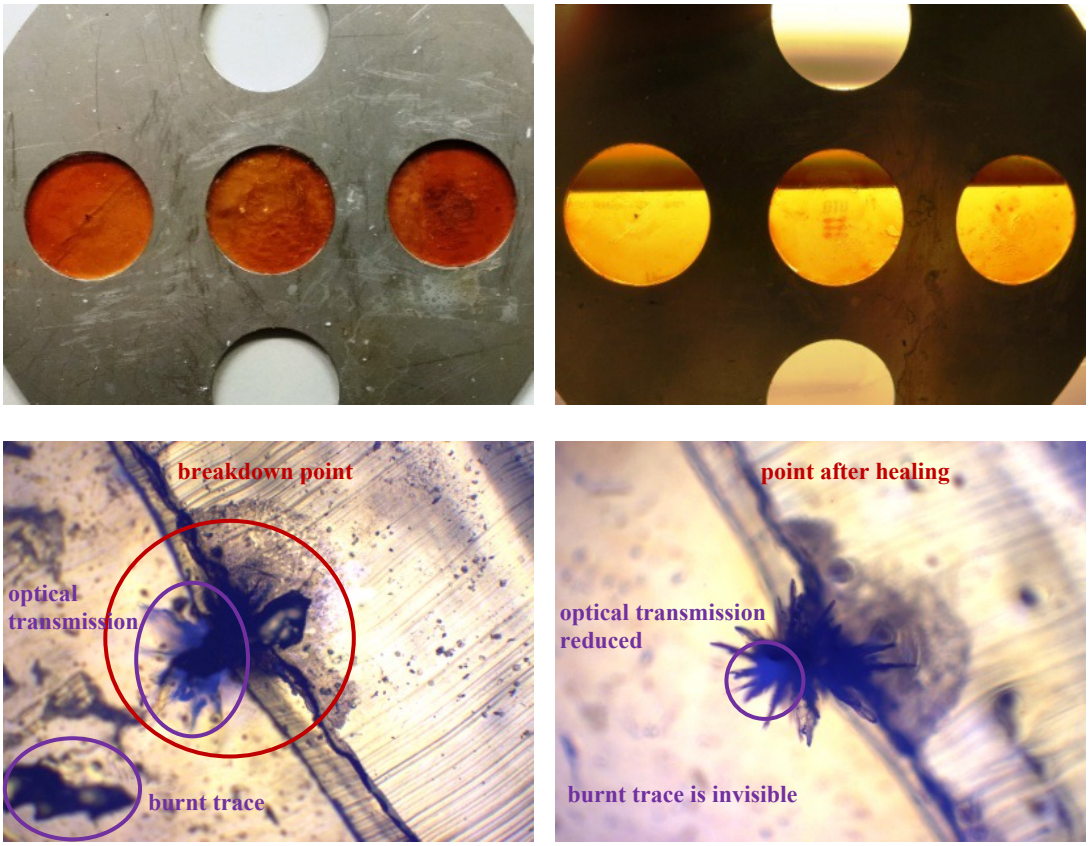

Figure 7. Pictures illustrating the samples before and after the self-healing upon electrical breakdown. Left figures are samples immediately after application of destructive electrical field. Right figures are after self-healing. The burnt area remains dark but the ionic network repairs the void.

4. CONCLUSION

Interpenetrating networks from commercial elastomers and ionic networks constitute a promising path for creating high-permittivity elastomers with self-repairing properties, both upon mechanical rupture but also upon electrical breakdown. The interpenetrating networks were furthermore shown to possess very good properties at low frequencies.

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